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Assessment of Natural Radionuclides in Sea Water and Associated Health Risks

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Abstract
Assessment of natural radionuclides in sea water and associated health risks have been done using NaI (TI) detector as a stable and reliable device for analysis of radionuclides. The result of activity concentration of radionuclides (^{40}K, ^{238}U and ^{232}Th) in sea water showed that ^{40}K has the highest value 180.40 Bql^{-1}. The lowest value was recorded by ^{232}Th due to it's low solubility in water. The percentage mean activity concentration of radionuclides in sea water are; 85%, 11% and 4% for ^{40}K, ^{238}U and ^{232}Th respectively. The absorbed dose range between 0.473 $nGyh^{-1}$ to 16.727 Gyh^{-1}. The comparison of the results with world standard showed that the calculated absorbed dose, annual effective dose for different age categories and excess lifetime cancer risk value of the identified radionuclides are within the world recommended safe value. However, the annual effective dose of different age categories showed that the annual effective dose of infants is higher than the value in children, while the adults recorded the lowest value. Therefore, the study concluded that though, the environment has been impacted radiologically, but there are no immediate identified radiation health risks as a result of radionuclide activity concentration in sea water of the study area.

1. Introduction

Radionuclide contamination has been attracting much attention of researchers both nationally and internationally. Radioisotopes also, referred to as radioactive isotopes or radionuclides, are highly unstable atoms that possess excess nuclear energy. The presence of different types and quantities of radionuclides in every given space around man's 'environment' is traceable to types and intensity of man's interference with nature and natural causes due to interaction of radiation or energy with matter.

The interaction of energy with matter in the environment can give rise to different effects which will in turn affect man and other living, and non-living matter therein. The natural environment is comprised of different components such as air, soil and water and their various types. Radionuclides as radiation sources are considered as one of the main sources of environmental pollutants. Radionuclides undergo radioactive decay and as such are found in nature, and can be artificially produced by man (UNSCEAR, 2018). The radionuclides found in the environment may cause harmful effects to man and the environment due to the fact that they possess excess energy as a result of their instability. This instability is what give rise to radioactivity or radioactive decay. Radioactivity is a function of radionuclides in any given environment which in turn are emitted into the environment by the use of x-rays and other ionizing radiation sources (WHO, 2020).

Radiation sources that are used in industry and trade, and by oil and gas industries contribute to the quantity and activity concentration of radionuclides in a given environmental media, which can affect the environment and man as a result of their interaction with matter.

European Atlas of Natural Radiation EANR, (2023) revealed that drinking water may contain radionuclides that could present a risk to human health, but this risk is normally low compared to risk from microorganisms and chemicals. Usually, the radiation dose resulting from ingestion of radionuclides in drinking water is much lower than that received from other radiation sources. Naturally occurring radionuclides in drinking water usually result in radiation doses that are higher than those provided by artificially produced radionuclides and are therefore of greater concern (WHO, 2017, 2018: EANR 2023).
Natural waters contain both alpha and beta emitters in widely varying concentrations which give relatively small contributions to the total dose received from natural and artificial radioactivity. Alpha activity is mostly due to dissolved uranium isotopes (^{234}U, ^{235}U and ^{238}U) and ^{226}Ra. The main source of gross beta activity in waters is ^{40}K and short-lived daughters of ^{238}U and ^{234}Th (Forte et al., 2007: EANR 2023). Natural radionuclides can be found in ground and surface water as a result of either natural processes (for example, reaction with aquifer minerals and absorption from the soil) or technological processes involving naturally occurring radioactive materials (such as mining and processing of mineral sands or phosphate fertilizer production) (WHO, 2017).
Anthropogenic sources of radioactivity in water include trans-uranium products like, ^{14}C, ^{3}H, ^{90}Sr and ^{131}I and other gamma emitters, released in controlled or authorized quantities from nuclear installations, medical or industrial facilities, or from past bomb-testing or nuclear accidents. The dominant radionuclides in ground and surface water that can pose a potential health hazard under natural conditions are ^{210}Po, ^{210}Pb, ^{210}Bi, ^{222}Rn, ^{226}Ra and ^{228}Ra in the ^{232}Th series (WHO, 2017).
Priasetyono *et al*., (2021) investigated natural and anthropogenic radionuclide concentrations in the Wakatobi and Kendari seas using a Gamma spectrometer (HPGe). The activity concentration of ^{137}Cs in both Wakatobi and Kendari range from BDL to 1.09 Bq m $^{-3}$. The activity concentration of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K range from 10.45 to 36.35 l^{-1}, 11.45 to 43.65 Bq l^{-1}, and 46.6 to 325 Bq l^{-1} respectively. The study concluded that the monitoring results showed that both natural and anthropogenic; (^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs) indicate that they are within the world acceptable values and do not exceed the safe limit.

2. Materials and Methods

2.1 Study Area

Nigerian Port authority Onne Port is located in Eleme along Ogu creek, Rivers state. The sea in Onne port as a part of Ogu River has harbour where ship berth under high level of security to offload containers, which are afterwards scanned with X-ray scanner. The sea in Onne port complex serves as a means of transportation for oil and gas industries, logistics, chemical and other allied industries. The port is strategically located in one of the largest Oil and Gas Free Zone in the world that is supporting exploration and production for Nigerian economic activities. The free zone provides a logistics oil service Centre for the oil and gas industry in Nigeria both onshore & offshore. It also provides easy access to the entire West African & Sub-Sahara oil fields. Onne port is a multipurpose cargo port and accounts for over 65% of the export cargo through the Nigerian sea port. There are multiple operations that are carried out in the port, in addition to oil & gas operations. Such numerous operations include general cargoes, oil well equipment, bulk cargoes (wet & dry), containerized cargoes and logistics services are provided to companies and customers or tenants. Onne port is highly industrialized with modern facilities and equipment, and enough land for development. The land area is about 2,538.115 hectares. The port is equipped with the biggest harbour mobile cranes (Liebherr 600) in Africa and with lifting capacity of 208 metric tons & 220 Gmk 5220 grove twin cranes that has the capacity to lift a single load with 300 tons (NPA 2023).

There are heavy industrial activities (oil exploration) due to the presence of crude oil (petroleum) in the area.
The state's geography is characterized by numerous rivers, including the **Bonny River**. These waterways play a significant role in the state's transportation and commerce. These numerous rivers, including the Bonny River flow through onne. The onne sea port which is located in rivers State with Its capital and largest city, Port Harcourt, and it is economically important as the centre of Nigeria's oil and gas industry (Okeowo, 2022). Rivers State is bounded on the South by the Atlantic Ocean, to the North by Imo, Abia and Anambra States, to the East, Akwa Ibom State, and to the West by Bayelsa and Delta states.
Rivers State is on Nigeria's coastline and has a number of seaports. Prior to the discovery of oil in Rivers State, Agriculture was the mainstay of its economy. About 39 % of the state's total land mass is suitable for crop cultivation. Basically, the vegetation is Tropical Rainforest and Monsoon. Rivers State also has a lucrative fishing industry, (NDBM, 2024).
Rivers State is the 26th largest state by area in Nigeria. The state is made up of different zones of land surface and water zones, this is further divided into three zones namely; freshwater swamps, mangrove swamps, and coastal sand ridges (NDBM, 2024).
However, the freshwater zone extends northwards from the mangrove swamps, and the land surface is generally less than 20 meters above sea level (NDBM, 2024).

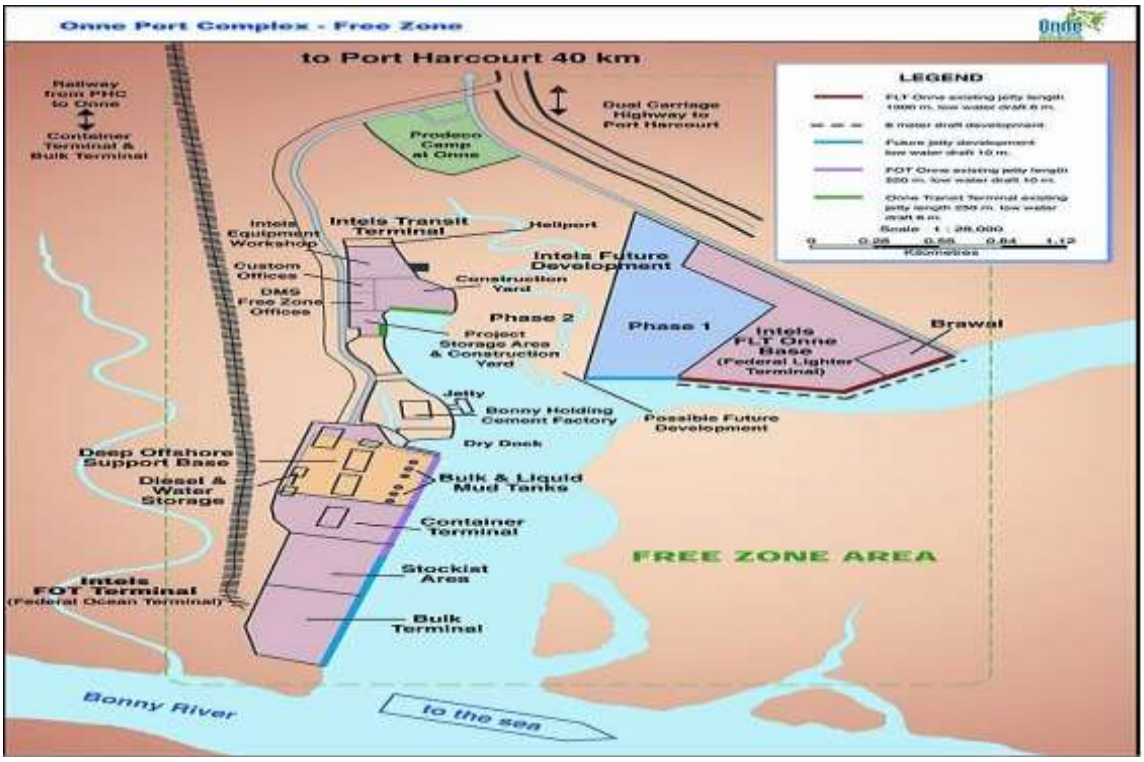

Figure 1: Map of Onne Sea Port Complex (Source: JEES; Ononugbo, C.P. *et al*, 2015)

2.2 Method of Data Collection

The method of data collection is observation method. The sea water samples were collected in white transparent plastic containers. Before the collection of water samples, the plastic containers were rinsed with the water which the containers were filled with afterwards. The water samples were collected at various distances from each sampling point. The collected sea water samples were acidified after collection.

3. Radiological Risk Parameters

3.1 Radium Equivalent (Ra_{eq})

In order to effectively estimate the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, an index known as radium equivalent activity (Ra_{eq}) has been presented. This is to enable a single index to define the radiation hazards associated with mixture of the radionuclides. Ra_{eq} was calculated from equation (1) with an assumption that 370 $Bqkg^{-1}$ of ^{226}Ra, 259 $Bqkg^{-1}$ of ^{232}Th and 4810 $Bqkg^{-1}$ of ^{40}K produce the same gamma-ray dose rate (Ravisankar *et al.*, 2014; Kolo *et al*., 2015).

$$Ra\_eq = AU + 1.43\ ATh + 0.077\ AK \quad (1)$$

Where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively.

Both external and internal gamma dose from exposure to radon and its daughter nuclides are related to Ra_{eq} (Ravisankar *et al.*, 2015: UNSCEAR, 2018).

3.2 Absorbed Dose Calculation

The absorbed dose rate (D) which is a measure of the amount of energy from an ionizing radiation deposited in a mass of some material or tissue (small unit of Gray) was calculated. Contributions from other radionuclides such as ^{137}Cs, ^{131}I and ^{90}Sr to total dose from environmental background were neglected in this calculation since they were assumed to be very insignificant. The absorbed dose rates D ($nGyh^{-1}$) was calculated using equation 2, (UNSCEAR,2000;UNSCEAR, 2010; Jibiri and Okeyode , 2012):

$$D\ (nGyh^{-1}) = (0.0090 A_U + 0.0070 A_{Th} + 0.028 A_K) \quad 2$$

Where A_U, A_{Th}, and A_K are the activity concentration of ^{238}U, ^{232}Th, and ^{40}K respectively in Bq l^{-1}

The dose conversion factors 0.0090, 0.0070, and 0.028 are expressed in ($nGy.h^{-1}Bqkg^{-1}$) as provided in the UNSCEAR, 2018 report.

3.3 Annual Effective Dose Equivalent Calculation

Annual effective dose rate is a measure of radiation risk associated with the air absorbed dose rate which represents the dose quantity of the stochastic health effects of low-level radiation on the human tissue. AEDE was calculated from D taking into cognizance two principal conversion factors provided by (UNSCEAR, 2000).

$$AED_{mSv/y} = A_c \times A_i \times C_f \quad 3$$

Where;

A_c is the activity concentration of the radionuclide in Water (Bq L^{-1}),

A_i is the consumption per annum or annual intake of water and

C_f is the ingested dose conversion factor for radionuclides ($SvBq^{-1}$), which varies for different radionuclides and the age of individual.

Equation (3) above is used to calculate annual effective dose equivalent using the activity concentration of radionuclides in sea water. The annual effective dose values were calculated for three different age groups (0-1yr, 7-12yr, >17yr) i. e infants, Children and Adults. The annual intake of water for the three different age categories is extracted from the publication of World Health Organization (WHO, 2003).

3.4 Committed Effective Dose

Committed effective dose is stochastic health risk due to an intake of radioactive material in the human body. The committed effective dose is a measure of the total effective dose received over an average life time of 50 years. However, the committed effective dose can only be calculated for adults.

$$CED = 50 \times D \quad (4)$$

Where D is the total effective dose to an individual.

3.5 Excess Lifetime Cancer Risk (ELCR)

The probability of occurrence of cancer in any given population for a given lifetime exposure is measured by the excess lifetime cancer risk (ELCR). ELCR was calculated from the estimated AEDE using the equation 2.14 (Qureshi *et al.*, 2014):

$$ELCR = AEDE \times DL \times RF \quad (5)$$

Where; DL is the life expectancy of 56 years and RF is risk factor given to be 0.05 Sv^{-1} for stochastic effects (Taskin *et al.*, 2009).

Table 1: Radionuclides and their Ingested Dose Conversion Factor for Different Age Categories (IAEA, 1996)

Radionuclide	Age Categories		
	Infant (0—1) yr Sv/Bq	Children (7— 12) yrs Sv/Bq	Adult >17yrs Sv/Bq
^{40}K	6.2E-8	1.3E-8	6.2E-9
^{238}U	4.7E-6	8.0E-7	2.8E-7
^{232}Th	4.6E-6	2.9E-7	2.3E-7

Table 2: Annual Water Consumption for Different Age Categories (WHO, 2003)

Age Categories	Annual Water Consumption (Lyr^{-1})
0—1	200
7—12	350
>17	730

4. Result

Table 3: Activity Concentration of Radionuclides (^{40}K, ^{238}U and ^{232}Th) in Sea Water Samples

S/N	SAMPLE CODE	K-40 (Bq/l)	U-238 (Bq/l)	Th-232 (Bq/l)
1.	RW1	180.40±8.70	16.98±1.28	1.22±0.07
2.	RW2	169.19±8.47	10.56±1.31	0.40±0.02
3.	RW3	0.90±0.05	BDL	0.008±0.0005
4.	RW4	161.38±8.13	8.83±1.04	8.78±0.51
5.	RW5	32.14±1.63	7.78±0.96	BDL
6.	RW6	44.28±2.14	12.19±0.95	2.19±0.12
7.	RW7	36.92±1.85	8.38±1.01	2.32±0.13
8.	RW8	72.95±3.51	3.20±0.38	1.47±0.09
9.	RW9	37.62±1.90	13.26±1.45	9.43±0.54
10	RW0	72.95±3.51	13.44±1.06	3.98±0.22
	Mean	**80.87±3.99**	**9.46±0.94**	**2.98±0.17**

Table 4: Estimated Absorbed Dose, Annual Effective Dose and Excess Lifetime Cancer Risk due to Radionuclides in Sea Water

S/N	SAMPLE CODE	(Bq/l) ^{238}U	^{232}Th	^{40}K	Ra_{eq}	D ($nGyh^{-1}$)	AEDE ($mSvy^{-1}$)	ELCR $\times 10^{-3}$
1	RW1	16.98	1.22	180.4	32.6154	16.01756	0.0982197	0.35089
2	RW2	10.56	0.4	169.2	24.15963	12.23044	0.0749971	0.3437689
3	RW3	BDL	0.01	0.9	0.08074	0.472928	0.0029	0.2624897
4	RW4	8.83	8.78	161.4	33.81166	16.72744	0.1025727	0.01015
5	RW5	7.78	BDL	32.14	11.68478	5.41	0.0331741	0.3590043
6	RW6	12.19	2.19	44.28	18.73126	8.62418	0.0528835	0.1161094
7	RW7	8.38	2.32	36.92	14.54044	6.75624	0.0414293	0.1850922
8	RW8	3.2	1.47	72.95	10.91925	5.55842	0.0340842	0.1450024
9	RW9	13.26	9.43	37.62	29.64164	13.61094	0.0834623	0.1192948
10	RW0	13.44	3.98	72.95	24.74855	11.6128	0.0712097	0.292118
MEAN		**10.5**	**3.31**	**80.9**	**20.09334**	**9.70209**	**0.059493**	**0.218392**

Table 5: Annual Effective Dose for Different Age Categories due to Activity Concentration of Radionuclides in Sea Water, and Committed Effective Dose for Adults

Age Categories	Radionuclide	Mean Activity Concentration (Bq/l)	Radionuclide Ingestion Dose convertion factor	Annual Water Consumption (l/yr)	AED (mSv/yr)	CED (mSvyr-1)
Infants	^{40}K	80.87	6.20E-08		1.00E-03	
	^{238}U	9.46	4.70E-06		8.89E-03	
	^{232}Th	2.98	4.60E-06	200	2.74E-03	
					1.26E-02	
Children	^{40}K	80.87	1.30E-08		3.68E-04	
	^{238}U	9.46	8.00E-07	350	2.64E-03	
	^{232}Th	2.98	2.90E-07		3.02E-04	
					3.31E-03	
Adults	^{40}K	80.87	6.20E-09		3.66E-04	
	^{238}U	9.46	2.80E-07	730	1.93E-03	0.90
	^{232}Th	2.98	2.30E-07		5.00E-04	
					2.80E-03	

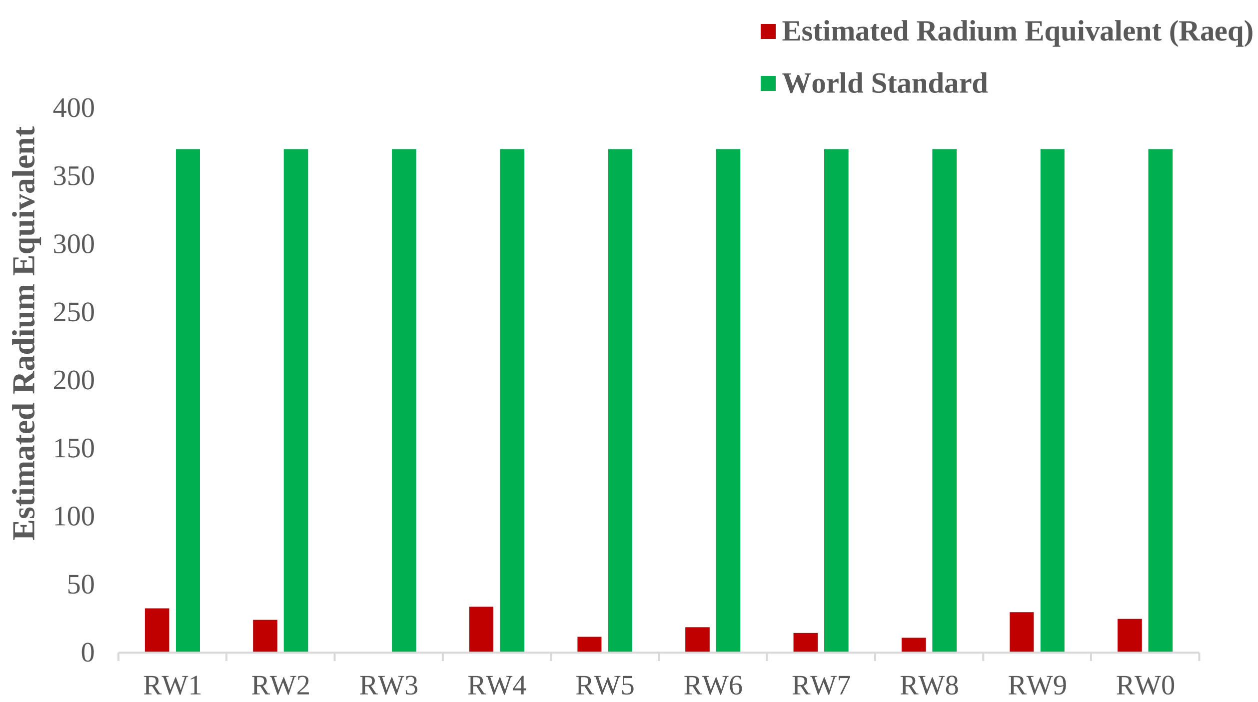

Figure 2: Comparison of Estimated Radium Equivalent of Radionuclides in Sea Water Samples with World Standard

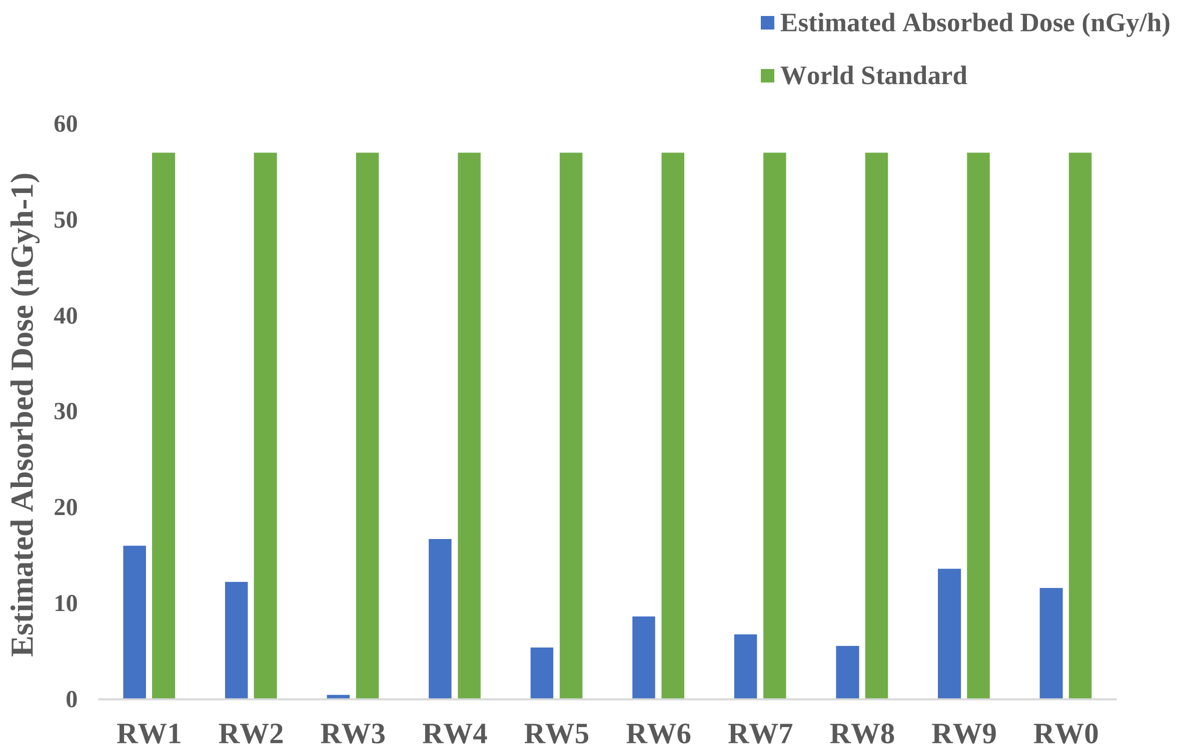

Figure 3: Comparison of Estimated Absorded Dose of Radionuclides in Sea Water with World Standard

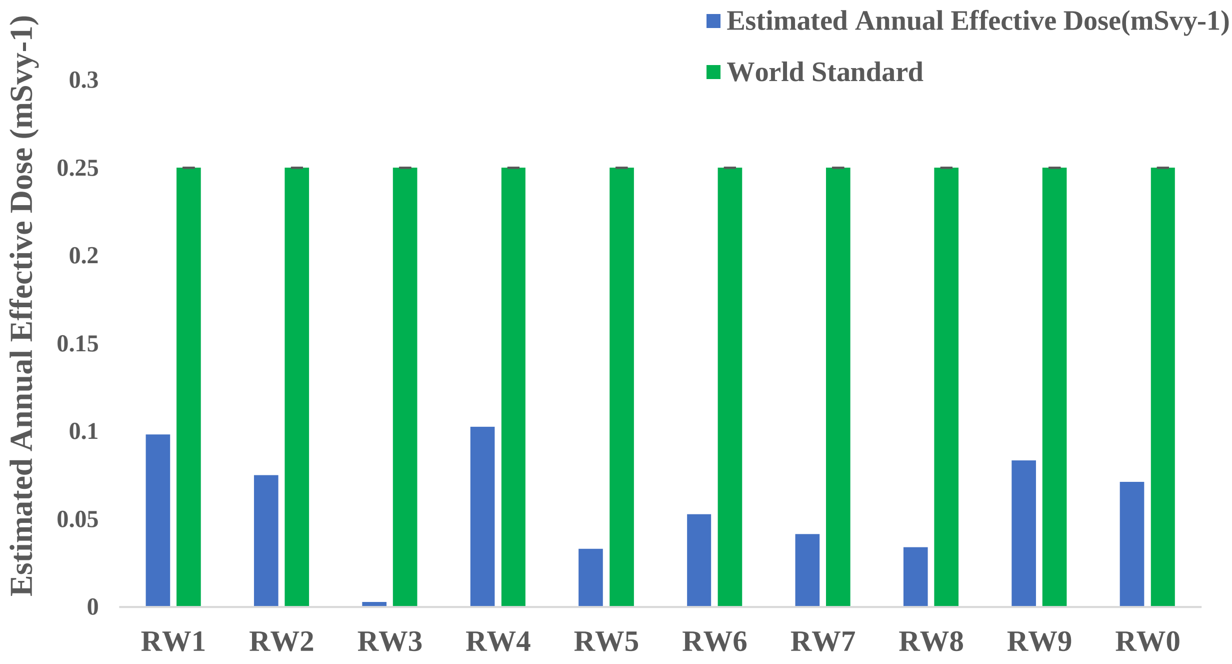

Figure 4: Comparison of Estimated Annual Effective Dose of Radionuclides in Sea Water with World Standard

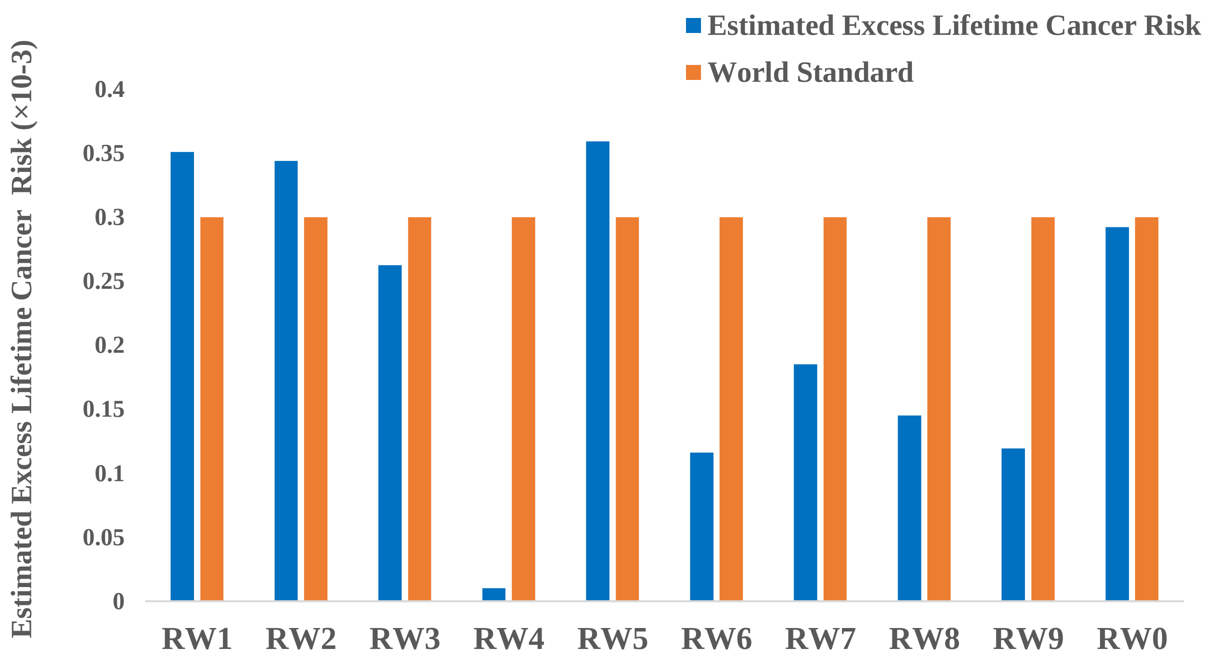

Figure 5: Comparison of Estimated Excess Lifetime Cancer Risk of Radionuclides in Sea Water with World Standard

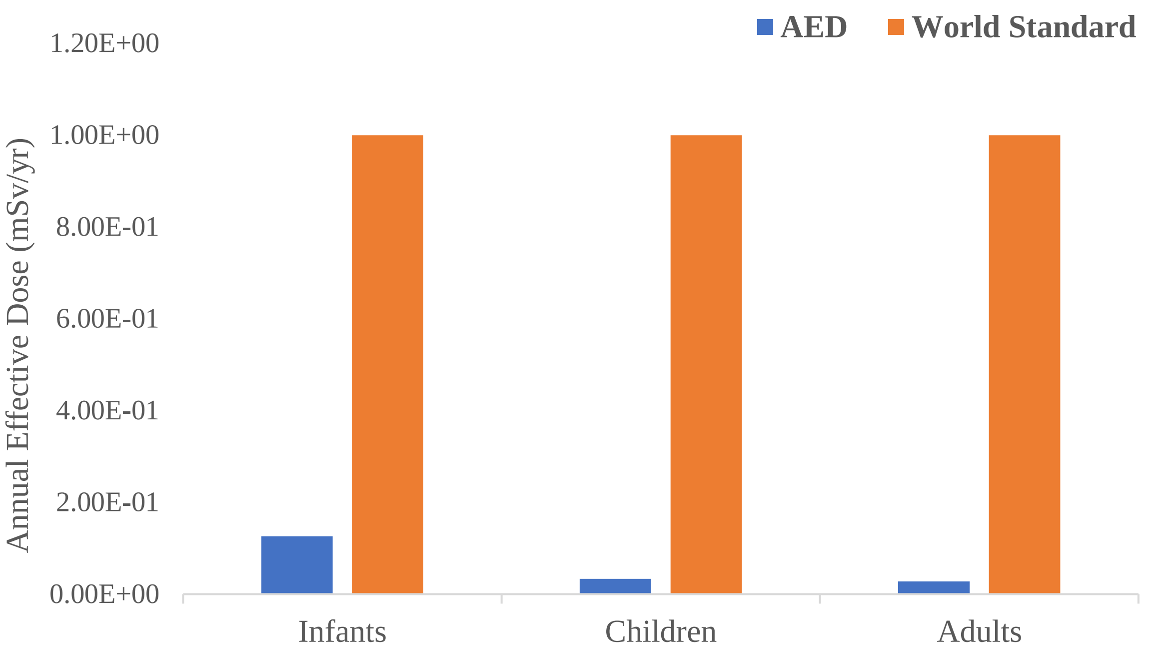

Figure 6: Comparison of Annual Effective Dose of Different Age Categories with World Standard

5. Discussion

The activity concentration of radionuclides in the sea water samples showed that activity concentration of ^{40}K, ^{238}U and ^{232}Th range between 180.40±8.70 Bql^{-1} to 0.90±0.05 Bql^{-1}, 16.98±1.28 Bql^{-1} to BDL and 9.43±0.54 Bql^{-1} to BDL respectively. While the mean activity concentration of ^{40}K, ^{238}U and ^{232}Th in the sea water samples is 80.88 Bql^{-1}, 10.51 Bql^{-1} and 3.31 Bql^{-1} respectively. The activity concentration of radionuclides in sea water samples showed that ^{40}K recorded the highest value of activity concentration, followed by ^{238}U, while ^{232}Th recorded the lowest activity concentration value. Thus, the percentage of mean activity concentration of radionuclides in sea water samples are; 85%, 11% and 4% for ^{40}K, ^{238}U and ^{232}Th respectively. This highest activity concentration of ^{40}K compared to other radionuclides as reported in this study is similar to the results obtained by Nimat *et al*, (2017), and Agyeman *et al*, (2019) in their studies. However, the mean values of activity concentration of radionuclides in this study are higher than the values recorded by the aforementioned studies. This high value is due to the use of linear accelerator (LINAC) in the study area which increases radiation burden of the environment therein. The effective activity concentration of radionuclides in sea water samples range between 34.0491 Bql^{-1} to 0.08698 Bql^{-1}, with a mean value of 20.3707 Bql^{-1}.

The range and mean value of estimated radium equivalent of radionuclides in sea water samples is between 33.81166 to 0.08074 Bql^{-1}, and 20.09334 Bql^{-1} respectively. Samples RW3 and RW4 recorded the lowest and highest values of radium equivalent due to radionuclides in water samples. The estimated radium equivalent value due to radionuclides in all the water samples is within the world recommended acceptable limit, this result is similar to the findings of Agyeman *et al*, (2019).

The absorbed dose of radionuclides in sea water samples range between 16.72744 $nGyh^{-1}$ to 0.472928 $nGyh^{-1}$, with a mean value of 9.70209 $nGyh^{-1}$. Samples RW3 and RW4 has lowest and highest values of radionuclide absorbed dose respectively. The values of absorbed dose in all sea water samples are acceptable, hence, within the world standard limit by United Nations Scientific committee on the Effects of Atomic Radiation (UNSCEAR). This result is in agreement with the result of Saif *et al*, 2017.
The annual effective dose equivalent (AEDE) of radionuclide in sea water range between 0.0029 to 0.1026 $mSvy^{-1}$ with a mean value of 0.0595 $mSvy^{-1}$. The calculated values of annual effective dose equivalent of the sea water are within the world acceptable standard value. The calculated values of excess lifetime cancer risk of radionuclide in sea water range between (0.01015 to 0.35089) $\times 10^{-3}$ with a mean value of 0.218392. About 30% of the samples have excess lifetime cancer risk values that are above the recommended world standard values.
The annual effective dose for infants is higher than the annual effective dose for children and adults when compared with different age categories. This high level of annual effective dose of radionuclide in infants is due to the radiation burden of the environment of the study area, and their ability to absorb more radiation due to the high radiosensitivity of their organs. The annual effective dose for the 3 basic age categories (Infants, children and adults) that are compared with world standard indicates that the values are all within the world acceptable value by National Commission on Radiation Protection (NCRP). This high level of annual effective dose in infants compared to other age categories is in agreement with the result of Ibikunle *et al*., 2018.
The committed effective dose for adults is within the recommended acceptable world value.

6. Conclusion

Assessment of natural radionuclides in sea water and associated health risks has been carried out using 10 sea water samples collected from the sea within Onne sea Port complex. The mean activity concentration of the radionuclides ^{40}K, ^{238}U and ^{232}Th are; 80.9 Bql^{-1}, 10.5 Bql^{-1} and 3.31 Bql^{-1} respectively. While the mean absorbed dose of the radionuclides is, 9.70209 $nGyh^{-1}$. The effective activity concentration of radionuclides (^{40}K, ^{238}U and ^{232}Th) in sea water are not dangerous. The values of radium equivalent, absorbed dose and annual effective dose equivalent of the radionuclides in sea water are within the world acceptable values.
These moderate or safe values of radionuclides recorded in the sea water are due to the dispersion nature of sea water and the adsorption of minerals within the medium. This is similar to the conclusion from a study by Priasetyono *et al* 2021.
The activity concentration, absorbed dose and radium equivalent of radionuclides in the sea water are within the world acceptable standard limit. The values of annual effective dose for different age categories due to the identified radionuclides in sea water are low, though within the world limit, but the value in infants is higher than in children and adults. However, ^{40}K radionuclide has the highest value of activity concentration compared to other radionuclides in the sea water. As such, the activity concentration level, and radiological health risks of radionuclides in Onne sea water do not pose an immediate radiation health risk.

7. Recommendation

Further research should be carried out using the aquatic animals and organisms from the sea water, and the result should be compared with other sea from other regions.

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